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Abstract. Within the scope of the Accelerator Driven System (ADS) concept for nuclear waste management applications, the burnup uncertainty estimates due to uncertainty in the activation cross sections (XSs) are important regarding both the safety and the efficiency of the waste burning process. We have applied both sensitivity analysis and Monte Carlo methodology to actinides burnup calculations in a lead-bismuth cooled subcritical ADS. The sensitivity analysis is used to identify the reaction XSs and the dominant chains that contribute most significantly to the uncertainty. The Monte Carlo methodology gives the burnup uncertainty estimates due to the synergetic/global effect of the complete set of XS uncertainties. These uncertainty estimates are valuable to assess the need of any experimental or systematic re-evaluation of some uncertainty XSs for ADS.

INTRODUCTION

Transmutation techniques based on subcritical systems, such as ADS, are considered one promising option to contribute to the solution of the nuclear waste management. A typical ADS concept [1] consists of an 800 MWth fast core cooled by lead-bismuth eutectic in forced convection. The core is subdivided into 132 hexagonal fuel assemblies. Table 1 shows the initial actinide composition of the fuel.

TABLE 1. Initial fuel composition (wt%)

Isotope	(wt%)	Isotope	(wt%)	Isotope	(wt%)
^{234}U	0.77	^{240}Pu	31.73	^{242}Cm	0.00
^{235}U	0.19	^{241}Pu	7.53	^{243}Cm	0.05
^{236}U	0.28	^{242}Pu	14.54	^{244}Cm	5.22
^{238}U	0.00	^{241}Am	6.28	^{245}Cm	1.80
^{237}Np	4.61	^{242M}Am	0.28	^{246}Cm	1.27
^{238}Pu	6.90	^{243}Am	5.99	^{247}Cm	0.19
^{239}Pu	12.35				

In previous studies, the simulation of this ADS, its k_{eff} values, power distributions and isotopic composition evolution during burnup was performed using the EVOLCODE system. EVOLCODE [1] uses the ORIGEN2.1 code to predict the isotopic inventory with had-oc libraries for the burnup calculations.

Now, we investigate the performance capability of the activation code ACAB [2] both to predict the isotopic inventory and to perform a comprehensive sensitivity-uncertainty analysis for burnup calculations in ADS. The potential of our ACAB code has been proved in an extensive number of studies for fusion applications [2,3,6,7].

First, we have performed burnup calculations with ACAB for the ADS concept [3]. Using the same nuclear data libraries (decay data and XS) processed by EVOLCODE to ORIGEN, we have concluded that these activation codes have the same computation reliability to predict the actinides inventory. Moreover, we have evaluated the importance to treat all possible pathways for the formation of any particular nuclide relevant for ADS. We have used ACAB to treat all the neutron reactions, ranging from thermal region to 20 MeV (ORIGEN only treats 6 different neutron reactions) and the possible decay transitions from ground, first and second isomeric states. After 100 days of burnup, comparison of this isotopic inventory

calculation with the previous one has shown that the transmutation sequence ^{242M}Am(n,n)^{242}Am (not considered in ORIGEN) causes differences in the inventory of two important isotopes: ^{242M}Am(-1.8%) and ^{242}Cm(+1.3%).

Second, we have introduced in ACAB the capability to perform a comprehensive sensitivity-uncertainty analysis in burnup calculations for ADS [3]. In the next section, we demonstrate the applicability and the importance of these analyses.

SENSITIVITY AND UNCERTAINTY ANALYSIS

It is well known that the activation XS uncertainties remain high for some isotopes, and this fact may cause significant uncertainties of the isotopic inventory prediction. In this section, we calculate uncertainty estimates of the actinides inventory.

Uncertainties in Activation XSs for ADS

In this work, we have taken XS uncertainty data from the FENDL UN/A-2.0 and EAF2003/UN libraries [4]. The information included in these uncertainty files reefers to limits for the relative error in the XSs, denoted hereafter as Δ_{LIM} (in fact, the information included in these files is $\Delta^2_{j,LIM}$, where *j* is the energy group). Uncertainties $\Delta_{j,LIM}$ are provided in a three-energy group structure for non-threshold reactions and in one energy group for threshold reactions. It is assumed that errors in XSs included in an energy group are 100% correlated, and that the errors in the different energy groups are uncorrelated. The uncertainties values, $\Delta_{j,LIM,}$ are included in these libraries as three times the experimental standard deviation, that is $\Delta_{j,LIM} = 3*\Delta_{j,EXP}$. (Table 2)

TABLE 2. Experimental uncertainty values included in EAF2003/UN.

Reaction	Energy (eV)	$\Delta_{j,EXP}$ (%)
Pu240 (n,γ)	1.0E-05 - 1.0E-01	3.43
	1.0E-01 - 4.0E+03	3.56
	4.0E+03 - 2.0E+07	16.67
Am241 (n,γ)	1.0E-05 - 1.0E-01	3.33
	1.0E-01 - 1.5E+02	8.13
	1.5E+02 - 2.0E+07	16.67

In order to measure the importance of these uncertainties for the ADS neutron spectrum, we define the corresponding total relative error (Δ) as shown in Equation 1. In Table 3, columns 2 and 3, uncertainties for (n,γ) reactions of some minor actinides are estimated. These uncertainties remain higher than 10%, and for some isotopes (Am, Cm) significant differences between FENDL/UN and EAF2003/UN are found.

$$\Delta^2 = \sum_{j=1}^{3} \Delta^2_{j,EXP} \cdot \left(\overline{\sigma}_j \Big/ \overline{\sigma} \right)^2 \qquad (1)$$

where, $\overline{\sigma}_j$ is the weighted XS in one of the energy groups, and $\overline{\sigma}$ the effective XS.

The uncertainties provided by FENDL/UN and EAF2003/UN are the most complete uncertainty libraries for inventory uncertainty prediction. To judge the adequacy of these uncertainty data for ADS, we have processed the ENDF/B-VI covariances of neutron XSs (FILE 33) using the NJOY99.0 code (ERRORR/COVR modules) [5]. We have converted energy-dependent covariance information in ENDF format into multigroup form, and we have obtained an equivalent total relative error (Δ_{EXP}) using Equation 2. Only covariance data for ^{240}Pu and ^{241}Am are given in ENDF/B-VI.

$$\Delta^2 = \sum_{\substack{i \in n \\ j \in n}} \alpha_i \cdot \mathrm{cov}(i,j) \cdot \alpha_j \qquad (2)$$

where, ϕ_i is the ADS multigroup neutron flux, $\alpha_i = \phi_i/\phi_T$, with $\phi_T = \sum_{i \in n} \phi_i$ and *n* is the total number of energy groups (175 groups); the term *cov(i,j)* is the ENDF/B-VI multigroup covariance matrix.

Comparing these uncertainty evaluations with the uncertainties calculated by EAF2003/UN and FENDL/UN we observe that: i) for ^{240}Pu(n, γ) the uncertainty is quite similar (~10%), ii) for ^{241}Am(n,γ) the uncertainty is reduced to one-fourth (~5%).

TABLE 3. Actinide (n,γ) XS uncertainties (Δ %).

Isotope	FENDL/UN	EAF2003/UN	ENDF/B-VI
^{237}Np	16.33	16.33	-
^{238}Pu	14.68	14.76	-
^{239}Pu	12.69	12.91	-
^{240}Pu	9.87	9.97	10.87
^{241}Pu	15.92	15.92	-
^{242}Pu	13.08	13.21	-
^{241}Am	25.45	16.30	4.82
^{242M}Am	33.27	16.63	-
^{243}Am	25.01	16.02	-
^{242}Cm	15.24	30.59	-
^{243}Cm	16.60	33.23	-
^{244}Cm	13.66	27.35	-

ACAB Methodology and Application

In the activation calculation, the XS uncertainty information (Tables 2,3) can be used in two different ways: i) to be used in a sensitivity- uncertainty analysis, ii) to assign probability density functions (PDF) for each of the XSs involved in a problem.

Sensitivity-uncertainty analysis

This procedure is based on a first order Taylor series approach, and in the use of original algorithms to compute efficiently the partial derivatives. [6] The approximation to provide the nuclide composition N_i depending on activation XS uncertainties is:

$$N_i \approx N_i\big|_{\sigma_0} + \sum_j \left(\frac{\partial N_i}{\partial \sigma_j}\right)_{\sigma_0} \cdot \left(\sigma_j - \sigma_{0j}\right) \quad (3)$$

where $N_i\big|_{\sigma_0}$ is the nuclide composition without uncertainties, and the sum is over all XSs.

This approximation can be re-written as:

$$\frac{N_i - N_i\big|_{\sigma_0}}{N_i\big|_{\sigma_0}} \approx \sum_{j=1} \rho_{ij} \cdot \frac{\left(\sigma_j - \sigma_{0j}\right)}{\sigma_{0j}} \quad (4)$$

The sensitivity coefficient (ρ_{ij}) for the production of *nuclide-i* due to uncertainty in *XS-j*, is defined by:

$$\rho_{ij} = \frac{\sigma_{j0}}{N_i\big|_{\sigma_0}} \left(\frac{\partial N_i}{\partial \sigma_j}\right)_{\sigma_0} \quad (5)$$

We have developed two different algorithms to calculate the partial derivatives. Originally, we implemented in ACAB a method based on numerical integration [6]. Now, using a slight modification of the readily routines we have implemented a new method to compute the partial derivatives. (An example of this calculation is shown in Table 4).

Denoting $Y_j = \left(\frac{\partial N}{\partial \sigma_j}\right)$, the time derivative expression will be:

$$\frac{d}{dt}\left(Y_j\right) = A \cdot \left(Y_j\right) + \left(\frac{\partial A}{\partial \sigma_j}\right) \cdot N; \quad Y_j(0) = 0 \quad (6)$$

Then, we solve at the same time **t** the nuclide concentration **N** and the partial derivative $\mathbf{Y_j}$:

$$\frac{d}{dt}\begin{pmatrix} N \\ Y_j \end{pmatrix} = \begin{pmatrix} A & 0 \\ B_j & A \end{pmatrix} \cdot \begin{pmatrix} N \\ Y_j \end{pmatrix}; \quad \begin{pmatrix} N \\ Y_j \end{pmatrix}(0) = \begin{pmatrix} N_0 \\ 0 \end{pmatrix} \quad (7)$$

N is the nuclide composition at time t, **A** is the n-by-n matrix involving the XS and decay values, and $\mathbf{B_j = \partial A / \partial \sigma_j}$

The sensitivity coefficient (ρ_{ij}) provides a direct measure of the *XS-j* importance to the nuclide formation-i (a sensitivity of 1.0 means that a 1% change in *XS-j* will cause a 1% change in *nuclide-i*). ρ_{ij} is burnup dependent, and the error-amplification factor of nuclide prediction will increase with burnup.

TABLE 4. Sensitivity coefficients (ρ_{ij}) of different nuclides for the reaction ^{241}Am(n,γ), at 100 days of burnup.

Isotope	ρ_{ij}	Isotope	ρ_{ij}	Isotope	ρ_{ij}
Am241	-0.02	Pu238	0.01	Am242	0.97
Am239	-0.02	Cm243	0.09	Cm238	0.97
Am240	-0.02	Cm241	0.95	Cm240	0.97
Am238	-0.02	Cm239	0.96	Cm242	0.97
Pu237M	0.01				

This methodology is found practical for providing the isotopic inventory uncertainties due to the uncertainties of each reaction XS. The relative error on the number of atoms ($\Delta N_i/N_i$) is given by Equation 8.

$$\left(\frac{\Delta N_i}{N_i}\right)^2 = \sum_j \rho_{ij}^2 * \left(\frac{\Delta \sigma_j}{\sigma_j}\right)^2 \quad (8)$$

The most important limitation of the method is that it is impractical to deal with the synergetic/global effect of the uncertainties of the complete set of XSs. To overcome this limitation, an uncertainty analysis methodology based on Monte Carlo has been implemented in ACAB.

Monte Carlo Methodology

The Monte Carlo procedure is based on simultaneous random sampling of all the XSs involved in the problem [7]. Authors of the FENDL library have noted that when evaluating XSs, the quantity $\log(\sigma_{expt}/\sigma_{calc})$ was approximately normally distributed [4]. Consequently, it can be said that for any given XS σ_j we can define the random variable $\log(\sigma_j/\sigma_{j0})$ that follows a normal distribution with mean 0 and

variance $\Delta^2_{j,EXP}$. The value σ_{j0} is the best-estimate XS from the standard XS file (i.e.: FENDL /A-2.0), and $\Delta^2_{j,EXP} = \Delta^2_{j,LIM}/9$ ($\Delta^2_{j,LIM}$ is given in the uncertainty file i.e.: FENDL UN/A-2.0).

The probability distributions of isotopic inventory (and estimate of the confidence level) are obtained with this method. A 1000 histories sample size is found appropriate for this application. We have defined a relative error of the isotopic inventory E_N, where $E_N=(N-N_0)/N_0$ and N_0 is the isotopic inventory obtained in the standard activation calculations, i.e, using the standard XSs file.

The 95th percentile of the N distribution is defined by N_{95}. And, the 95th percentile of the distribution of E_N is defined by $E_{N,95}$ [$E_{N,95}=(N_{95}-N_0)/N_0$]. $E_{N,95}$ provides valuable information about the uncertainties. Table 5 gives $E_{N,95}$ for different important isotopes in the ADS. As we expected, $E_{N,95}$ depends on the burnup time. The isotopes with the maximum values of $E_{N,95}$ at 100 days of burnup are: ^{242M}Am and ^{242}Cm, 12.9% and 42.1%, respectively.

The XS responsible for the uncertainty in the prediction of ^{242}Cm is the reaction ^{241}Am(n,γ). This XS has a sensitivity coefficient of 0.97 (see table 4). Because of this effect and the high uncertainty differences between EAF2003/UN (16.3%) and ENDF/B-VI (4.82%) (see Table 4), it is worthwhile revising its XS uncertainty in EAF2003/UN.

TABLE 5. $E_{N,95}$ for the initial actinides in the fuel, at two different burnup time-steps, using EAF2003/UN.

Time (days)	^{234}U	^{235}U	^{236}U	^{238}U	^{237}Np	^{238}Pu
5	0.20	1.38	0.06	-0.22	0.37	0.31
100	0.90	9.33	0.50	0.40	0.78	1.71
	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	^{242m}Am
5	0.20	0.18	0.57	0.15	0.30	2.1
100	0.48	0.10	3.22	0.26	0.51	12.9
	^{243}Am	^{242}Cm	^{243}Cm	^{244}Cm	^{245}Cm	^{246}Cm
5	0.43	25.7	0.51	0.57	1.21	0.06
100	2.01	42.1	6.33	3.39	8.46	1.67

CONCLUSIONS

The ACAB code has been applied successfully to predict the isotopic inventory for ADS.

We have estimated and assessed the XS uncertainties in the most important actinides for ADS. A revision of the ^{241}Am(n,γ) uncertainty in EAF2003/UN has been proposed.

The suitability of the Monte Carlo methodology to deal efficiently with the synergetic/global effect of the uncertainties of the complete set of XSs on ADS is proved. The Monte Carlo methodology and the sensitivity analysis allow fully comprehensive uncertainty assessment of inventory calculations. The capability of ACAB to predict uncertainties in the inventory has been proved to be important regarding the ADS overall safety and the efficiency of the waste burning process.

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